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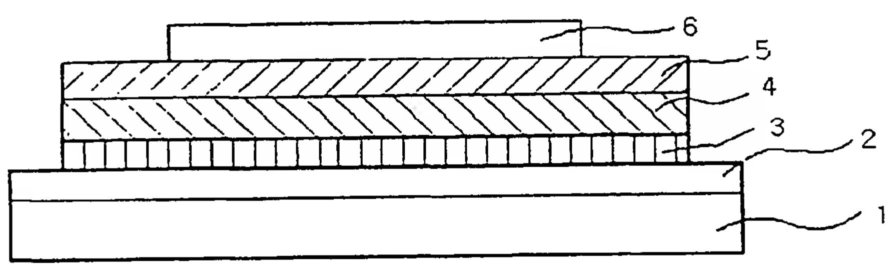
(54) Title: ORGANIC/POLYMER ELECTROLUMINESCENT DEVICE EMPLOYING SINGLE-ION CONDUCTOR



(57) Abstract: The present invention relates to organic/polymer electroluminescent devices employing single-ion conductors as the materials for an electron- or hole-injecting layer. The organic/polymer electroluminescent devices of the invention are improved in a sense that it employs an electron- or hole-injecting layer made of single-ion conductors in a conventional electroluminescent device which comprises: a transparent substrate; a semitransparent electrode deposited on the transparent substrate; a hole-injecting layer positioned on the semitransparent electrode; an electroluminescent layer made of organic luminescent material, positioned on the hole-injecting layer; an electron-injecting layer positioned on the electroluminescent layer; and, a metal electrode deposited on the electron-injecting layer. The organic/polymer EL devices of the invention have excellent EL efficiency and low turn-on voltage, which make possible their application to the development of high efficiency organic/polymer EL devices.

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ORGANIC/POLYMER ELECTROLUMINESCENT DEVICE EMPOLYING SINGLE-ION CONDUCTOR

BACKGROUND OF THE INVENTION

Field of the Invention

The present invention relates to organic/polymer electroluminescent devices employing single-ion conductors, more specifically, to organic/polymer electroluminescent devices employing single-ion conductors as an electron- or hole-injecting layer.

Description of the Prior Art

Electroluminescent("EL") device that emits light by applying an electric field to the device comprises ITO substrate, EL material and two electrodes. To improve the EL efficiency, the device is provided with a hole-injecting layer between the ITO electrode and EL material, an electron-injecting layer between EL material and the counter metal electrode, or both layers. As the EL material that plays a crucial role in the device, organic polymer/inorganic hybrid nanocomposite employing insulating inorganic materials, such as SiO_2 and TiO_2 that help the transport of electric charges, has been developed and put to the practical use (see: S. A. Carter, Applied Physics Letters, 71:1145, 1997; L. Gozano, Applied Physics Letters, 73:3911, 1998).

In the meantime, studies on the hole- or electron-injecting layer have been actively performed to improve the EL efficiency, mainly by way of inserting ionomers as the electron-injecting layer (see: Hyang-Mok Lee et al., Applied Physics Letters, 72, 2382, 1998). However, it cannot be a basic solution to improve the EL efficiency because the movement of ions is restricted in the ionomers, which

naturally limits electron-injection. As an alternative means for efficient electron-injection, an electron-transporting layer rather than the electron-injecting layer, was proposed in the art, which utilizes the materials that well transport electrons and have high affinity to the electrons. Several methods that utilize inorganic nanoparticles, 2-(4-biphenylyl)-5-(4-tert-butylphenyl)-1,3,4-oxadiazole(PBD), or metal chelate complexes have been presented until now(see: USP 5,537,000; USP 5,817,431; USP 5,994,835). However, these methods have not been realized in practical use due to the low EL efficiency or the difficulties confronted in the thin film deposition process.

Under the circumstances, there are strong reasons for developing and exploring a material that can be used as the hole- or electron-injecting layer to improve the EL efficiency while employing the convenient thin-film deposition process such as a spin-coating method.

SUMMARY OF INVENTION

The present inventors made an effort to develop a material that can improve the EL efficiency with convenient thin-film deposition process, and discovered that EL devices employing single-ion conductors as an electron- or hole-injecting layer show a highly improved EL efficiency.

A primary object of the present invention is, therefore, to provide EL devices employing single-ion conductors as an electron- or hole-injecting layer.

BRIEF DESCRIPTION OF THE DRAWINGS

The above, the other objects and features of the invention will become apparent from the following descriptions given in conjunction with the accompanying drawings, in which:

Figure 1 is a schematic diagram showing a cross-sectional view of an organic/polymer EL device employing single-ion conductors of the present invention.

Figure 2 is a graph showing the EL efficiency of an organic/polymer EL device employing a single-ion conductor as the electron-injecting layer, an organic/polymer EL device employing an ionomer as the electron-injecting layer, and an organic/polymer EL device without the electron-injecting layer.

<Explanation of major parts of the drawings>

1: transparent substrate
2: semitransparent electrode
3: hole-injecting layer
4: electroluminescent layer
5: electron-injecting layer
6: metal electrode

DETAILED DESCRIPTION OF THE INVENTION

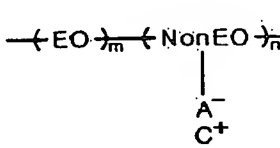
The organic/polymer EL device of the invention is improved in a sense that it employs electron- or hole-injecting layer made of single-ion conductors in a conventional EL device which comprises: a transparent substrate; a semitransparent electrode deposited on the transparent substrate; a hole-injecting layer positioned on the semitransparent electrode; an emissive layer made of organic luminescent material, positioned on the hole-injecting layer; an electron-injecting layer positioned on the emissive layer; and, a metal electrode deposited on the electron-injecting layer. The transparent substrate includes glass, quartz or PET(polyethylene terephtalate), and the semitransparent electrodes includes ITO(indium tin oxide), PEDOT(polyethylene dioxythiophene) or polyaniline.

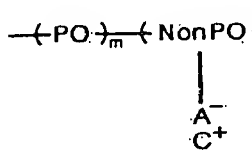
The organic EL material includes: emissive conjugated

polymers such as poly(para-phenylvinylene), poly(thiophene), poly(para-phenylene), poly(fluorene) or their derivatives; emissive non-conjugated polymers with side chains substituted with emissive functional groups such as anthracene; metal chelate complex of ligand structure such as emissive alumina quinone(Alq3); low molecular-weight emissive organic material (monomers or oligomers) such as rubrene, anthracene, perylene, coumarine 6, Nile red, aromatic diamine, TPD(N,N'-diphenyl-N,N'-bis-(3-methylphenyl)-1,1'-biphenyl-4,4'-diamine), TAZ(3-(4-biphenyl)-4-phenyl-5-(4-tert-butylphenyl)-1,2,4-triazole) or other emissive monomeric or oligomeric material of the derivative of those material; laser dyes such as DCM(dicyanomethylene)-2-methyl-6-(*p*-dimethylaminostyryl)-4*H*-pyran), and blends of poly(meta-methylacrylic acid), polystyrene and poly(9-vinylcarbazole) with above-mentioned emissive materials. And, aluminum, magnesium, lithium, calcium, copper, silver, gold, or an alloy thereof is preferably employed for the metal electrode.

As the single-ion conductors, the materials containing ether chains ($(-CH_2)_nO-$) such as polyethylene oxide or polypropylene oxide, and ionic groups such as SO_3^-, COO^-, I^-, or $(NH_3)_4^+$ in the main chains that form ionic bonds with counter ions such as Na^+, Li^+, Zn^{2+}, Mg^{2+}, Eu^{3+}, COO^-, SO^{3-}, I^-, or $(NH_3)_4^+$ are preferably employed.

In general, single-ion conductors are classified into single-cation conductors(see: general formula (I), general formula (II)) and single-anion conductors(see: general formula (III) and general formula (IV)).

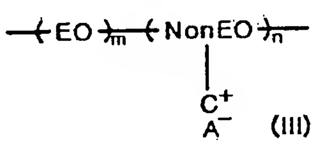

(I)

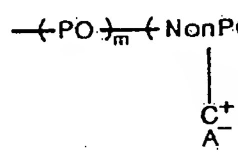

(II)

wherein,

EO represents ethyleneoxide;
NonEO represents non-ethyleneoxide;
PO represents propyleneoxide;
NonPO represents non-propyleneoxide;
A^- represents anion;
C^+ represents cation;
m+n=1; and,
n represents a real number more than 0 and less than 1.

As shown in the general formula (I) and the general formula (II), single-cation conductors contain ether chains ($(-CH_2)_nO-$) such as polyethyleneoxide or polypropyleneoxide in the main chains, and anionic groups such as SO_3^-, COO^-, or I^- in the main or side chains which form ionic bonds with metal ions such as Na^+, Li^+, Zn^{2+}, Mg^{2+}, or Eu^{3+}, or other organic ions such as $(NH_3)_4^+$ as the counter ion.


(III)


(IV)

wherein,

EO represents ethyleneoxide;
NonEO represents non-ethyleneoxide;
PO represents propyleneoxide;
NonPO represents non-propyleneoxide;
A^- represents anion;
C^+ represents cation;
m+n=1; and,
n represents a real number more than 0 and less than 1.

As shown in the general formula (III) and the general formula (IV), single-anion conductor contains ether chains ($(-CH_2)_nO-$) such as polyethyleneoxide or polypropyleneoxide

in the main chains, and cationic group such as $(NH_3)_4^+$ or $(-CH_2-)_nO^+$ in the main or side chains which form ionic bonds with anions such as SO_3^-, COO^-, or I^- as counter ion.

In the single-ion conductors descried above, the ether chain dissociates counter ions from the ions attached to the main chain and allows the ions to move much more freely. The EL intensity and the EL efficiency can be improved by employing the single-anion conductor as a hole-injecting layer or the single-cation conductor as an electron-injecting layer. However, the organic/polymer EL devices can be prepared to include either the hole-injecting layer or the electron-injecting layer to optimize the EL intensity and efficiency.

A preferred embodiment of the organic/polymer EL device of the present invention employing single-ion conductors is schematically depicted in Figure 1. The organic/polymer EL device employing single-ion conductors comprises a hole-injecting layer(3) that is prepared by spin-coating of the single-anion conductor on the ITO layer prepared by depositing the semitransparent electrode material(2) on the transparent substrate(1); an emissive layer(4) prepared by spin-coating of the organic emissive material on the hole-injecting layer(3); an electron-injecting layer(5) prepared by spin-coating of the single-anion conductor on the emissive layer(4); and, a metal electrode prepared by a thermal evaporation method using the metal such as Al, Mg, Li, Ca, Au, Ag, Pt, Ni, Pb, Cu, Fe, or their alloys on the electron-injecting layer(5).

As described above, when single-ion conductors are used in multi-layer EL devices, the conductivity is greater than 1×10^{-8} s/cm. The EL efficiency of the device is described in quantum efficiency (% photons/electrons), which indicates the number of photons per the number of electron injected in a limit of % probability. The EL external quantum efficiency (external quantum efficiency= externally emitted photons/injected electrons*100(%)) determined was between 0.5 and 2% photons/electrons, and the turn-on

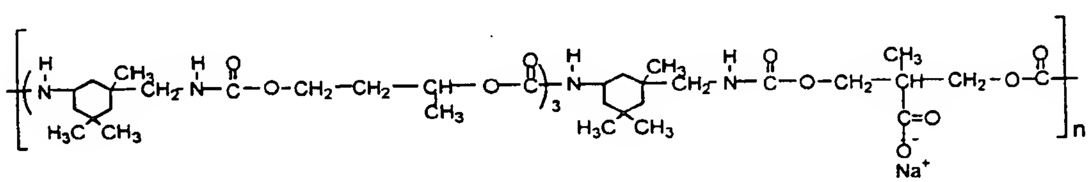
voltage for the emission was as low as 1.8V.

The present invention is further illustrated by the following examples, which should not be taken to limit the scope of the invention.

Example 1: Preparation of an organic/polymer EL device employing a single-cation conductor as an electron-injecting layer

A derivative of poly(para-phenylenevinylene), MEH-PPV (poly[2-methoxy-5-(2'-ethyl-hexyl)-*p*-phenylenevinylene]) was spin-coated on ITO substrate in 60 nm thickness as an EL material, and then a single-cation conductor with structural formula(I) below, which has Na^+ as a counter ion by ionic bond formation, was spin-coated in 15nm thickness on the the MEH-PPV layer. After that, an aluminum electrode was deposited in 100 nm thickness by a thermal evaporation method to give an organic/polymer EL device. The EL intensity was measured using a photodiode(818-UV) connected to an optical powermeter (Newport 1830-C) after applying a forward bias electric field. When EL efficiency against current density of the organic/polymer EL device was calculated by measuring current while applying voltage using Keithley 236 Source measurement unit, the turn-on voltage for emission of the organic/polymer EL device was 1.8V.

[Formula I]



Comparative Example 1: Preparation of an organic/polymer EL device without an electron-injecting layer

An organic/polymer EL device without an electron-injecting in Example 1, except that the spin-coating of a single-cation conductor was omitted, and EL efficiency against current was calculated.

Comparative Example 2: Preparation of an organic/polymer EL device employing an an ionomer as an electron-injecting layer

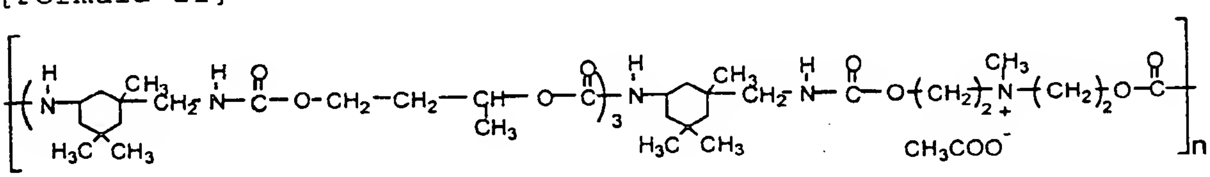
An organic/polymer EL device was fabricated in a similar manner as in Example 1, except that the known electron-injecting material, a SSPS ionomer (sodium sulfonated polystyrene) was used, and then EL efficiency against current was calculated to compared with the EL efficiencies in Example 1 and Comparative Example 1(see: Figure 2). Figure 2 depicts a graph comparing the EL efficiencies depending on the current densities of the organic/polymer EL devices in Example 1, Comparative Examples 1 and 2. In Figure 2, (△) represents the EL efficiency in case of employing a single-cation conductor as an electron-injecting layer, (○) represents the EL efficiency of the device employing an ionomer as an electron-injecting layer, and (□) represents the EL efficiency when the electron-injecting layer was not used. As shown in Figure 2, the EL efficiency of the invented organic/polymer EL device, employing a single-cation conductor as an electron-injecting layer, was improved by about 600 times as compared with that of not employing the electron-injecting layer, and by about 5 times compared with that of employing an ionomer as an electron-injecting layer. Further, the external quantum efficiency was calculated from the obtained results, for the invented organic/polymer EL device employing a single-cation conductor as an electron-injecting layer, which revealed that it was about 1%(photons/electrons), and for the organic/polymer EL device employing an ionomer as an electron-injecting layer, about

0.2%(photons/electrons), and for the organic/polymer EL device without the electron-injecting layer, about 0.004%(photons/electrons), which demonstrated that the organic/polymer EL device of the present invention is highly improved in terms of the EL efficiency by employing a single-cation conductor as an electron-injecting layer.

Example 2: Preparation of an organic/polymer EL device employing a single-anion conductor as a hole-injecting layer(1)

A single-anion conductor with the structural formula(II) below was spin-coated in 15nm thickness on the ITO anode substrate followed by spin-coating of the EL material, MEH-PPV in 100 nm thickness. And then, an aluminum cathode was deposited in 100 nm thickness by a thermal evaporation method to give an organic/polymer EL device. When the EL device was activated by applying a forward electric field, the turn-on voltage for emission of the organic/polymer EL device was 1.8V.

[Formula II]



Example 3: Preparation of an organic/polymer EL device employing a single-anion conductor as an hole-injecting layer(2)

An EL material, MEH-PPV was spin-coated on the ITO cathode substrate in 100nm thickness followed by spin-coating of a single-anion conductor with the structural formula(II) above 15nm in thickness. And then, an aluminum anode was deposited in 100nm thickness by a thermal evaporation method to give an organic/polymer EL device.

When the EL device was activated by applying reverse electric field, the turn-on voltage for emission of the organic/polymer EL device was 1.8V.

Example 4: Preparation of an organic/polymer EL device employing a single-anion conductor as a hole-injecting layer and a single-cation conductor as an electron-injecting layer

A single-anion conductor with the structural formula(II) above was spin-coated in 15nm thickness on the ITO substrate followed by spin-coating of the EL material, MEH-PPV in 100nm thickness. After the single-cation conductor with structural formula(I) was spin-coated in 15 nm thickness on the emissive layer, an aluminum electrode was deposited in 100nm thickness by a thermal evaporation method to give an organic/polymer EL device. The EL intensity was measured while activating the EL device by applying forward electric fields. The turn-on voltage for emission of the organic/polymer EL device was 1.8V.

As clearly described and demonstrated as above, the present invention provides organic/polymer EL devices employing single-ion conductors as an electron- or hole-injecting layer. The organic/polymer EL device of the invention is improved in a sense that it employs an electron- or hole-injecting layer made of single-ion conductors in the EL device which comprises: a transparent substrate; a semitransparent electrode deposited on the transparent substrate; a hole-injecting layer positioned on the semitransparent electrode; an emissive layer made of an organic emissive material, positioned on the hole-injecting layer; an electron-injecting layer positioned on the emissive layer; and, a metal electrode deposited on the electron-injecting layer. The organic/polymer EL devices of the invention have excellent EL efficiency and low turn-on voltage, which make possible their application to the

development of high efficiency organic/polymer EL devices.

Although the preferred embodiments of present invention have been disclosed for illustrative purpose, those who are skilled in the art will appreciate that various modifications, additions, and substitutions are possible, without departing from the spirit and scope of the invention as disclosed in the accompanying claims.

WHAT IS CLAIMED IS:

1. In an organic/polymer electroluminescent(EL) device which comprises: a transparent substrate; a semitransparent electrode deposited on the transparent substrate; a hole-injecting layer positioned on the semitransparent electrode; an emissive layer made of an organic EL material, positioned on the hole-injecting layer; an electron-injecting layer positioned on the emissive layer; and, a metal electrode deposited on the electron-injecting layer, the improvement comprising that single-ion conductors are employed for the hole-injecting layer and the electron-injecting layer.

2. The organic/polymer EL device of claim 1, wherein the transparent substrate is glass, quartz or PET(polyethylene terephtalate).

3. The organic/polymer EL device of claim 1, wherein the semitransparent electrode is lead oxide, ITO (indium tin oxide), doped polyaniline, doped Polypyrrole, doped polythiophene or PEDOT(polyethylene dioxythiophene).

4. The organic/polymer EL device of claim 1, wherein the organic EL material is emissive conjugated polymer, emissive non-conjugated polymer, emissive small organic (monomeric or oligomeric) material, poly(meta-methylacrylic acid), poly(styrene) or poly(9-vinylcarbazole).

5. The organic/polymer EL device of claim 4, wherein the emissive conjugated polymer is poly(*p*-phenylene vinylene), poly(thiophene), poly(*p*-phenylene), poly(fluorene), poly(arylenes), poly(arylene vinylene), polyquinoline, polypyrrole, polyaniline, polyacetylene or derivatives thereof.

6. The organic/polymer EL device of claim 4, wherein

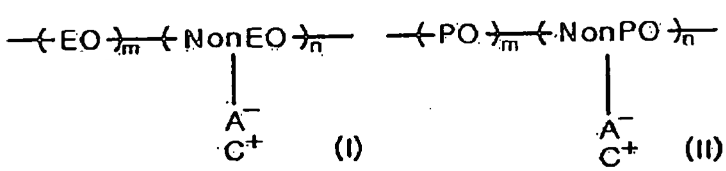
the emissive non-conjugated polymer is a polymer having non-conjugated main chains and side chains substituted with emissive functional groups.

7. The organic/polymer EL device of claim 4, wherein the emissive small organic (monomeric or oligomeric) material is alumina quinone(Alq3), rubrene, anthracene, perylenene, coumarine 6, Nile red, aromatic diamine, TPD(N,N'-diphenyl-N,N'-bis-(3-methylphenyl)-1,1'-biphenyl-4,4'-diamine), TAZ(3-(4-biphenyl)-4-phenyl-5-(4-tert-butylphenyl)-1,2,4-triazole), DCM(dicyanomethylene)-2-methyl-6-(*p*-dimethylaminostyryl)-4*H*-pyran), derivatives thereof.

8. The organic/polymer EL device of claim 1, wherein the metal electrode is made of aluminum, magnesium, lithium, calcium, copper, silver, iron, platinum, indium, palladium, tungsten, zinc, gold, lead or alloys thereof.

9. The organic/polymer EL device of claim 1, wherein the single-ion conductor is a single-cation conductor or a single-anion conductor.

10. The organic/polymer EL device of claim 9, wherein the single-cation conductor represented as a general formula (I) or (II) below, comprises ether chain ($(-CH_2)_nO-$) such as polyethylene oxide or polypropylene oxide in the main chain, and contains anions such as SO_3^-, COO^- or I^- in the main or side chains that form ionic bonds with counter ion such as Na^+, Li^+, Zn^{2+}, Mg^{2+}, Eu^{3+}, or $(NH_3)_4^+$:

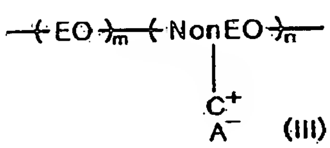


(I) (II)

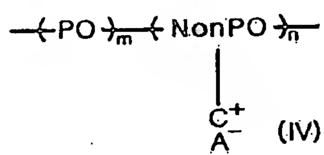
wherein,

EO represents ethyleneoxide;
NonEO represents non-ethyleneoxide;
PO represents propyleneoxide;
NonPO represents non-propyleneoxide;
A^- represents anion;
C^+ represents cation;
m+n=1; and,
n represents a real number more than 0 and less than 1.

11. The organic/polymer EL device of claim 9, wherein the single-anion conductor represented as a general formula (III) or (IV) below, comprises ether chain $((-CH_2)_nO-)$ such as polyethylene oxide or polypropylene oxide in the main chain, and contains cations in the main or side chains, such as $(NH_3)_4^+$ or $(-CH_2-)_nO^+$ that form ionic bonds with counter ions such as COO^-, SO_3^- or I^-:



(III)



(IV)

wherein,

EO represents ethyleneoxide;
NonEO represents non-ethyleneoxide;
PO represents propyleneoxide;
NonPO represents non-propyleneoxide;
A^- represents anion;
C^+ represents cation;
m+n=1; and,
n represents a real number more than 0 and less than 1.

12. An organic/polymer EL device which comprises:

a transparent substrate;

a semitransparent electrode deposited on the transparent substrate;

a hole-injecting layer made of single-anion conductors, positioned on the semitransparent electrode;

an emissive layer made of organic EL material, positioned on the hole-injecting layer;

an electron-injecting layer made of single-cation conductors, positioned on the emissive layer; and,

a metal electrode deposited on the electron-injecting layer.

13. An organic/polymer EL device which comprises:

a transparent substrate;

a semitransparent electrode deposited on the transparent substrate;

a electron-injecting layer made of single-cation conductors, positioned on the semitransparent electrode;

an emissive layer made of organic EL material, positioned on the electron-injecting layer;

an hole-injecting layer made of single-anion conductors, positioned on the emissive layer; and,

a metal electrode deposited on the hole-injecting layer.

14. An organic/polymer EL device which comprises:

a transparent substrate;

a semitransparent electrode deposited on the transparent substrate;

a hole-injecting layer made of single-anion conductors, positioned on the semitransparent electrode;

an emissive layer made of organic EL material, positioned on the hole-injecting layer; and,

a metal electrode deposited on the emissive layer.

15. An organic/polymer EL device which comprises:

a transparent substrate;

a semitransparent electrode deposited on the

transparent substrate;

a electron-injecting layer made of single-cation conductors, positioned on the semitransparent electrode;

an emissive layer made of organic EL material, positioned on the electron-injecting layer; and,

a metal electrode deposited on the electron-injecting layer.

16. An organic/polymer EL device which comprises:

a transparent substrate;

a semitransparent electrode deposited on the transparent substrate;

an emissive layer made of organic EL material, positioned on the semitransparent electrode;

an electron-injecting layer made of single-cation conductors, positioned on the emissive layer; and,

a metal electrode deposited on the electron-injecting layer.

17. An organic/polymer EL device which comprises:

a transparent substrate;

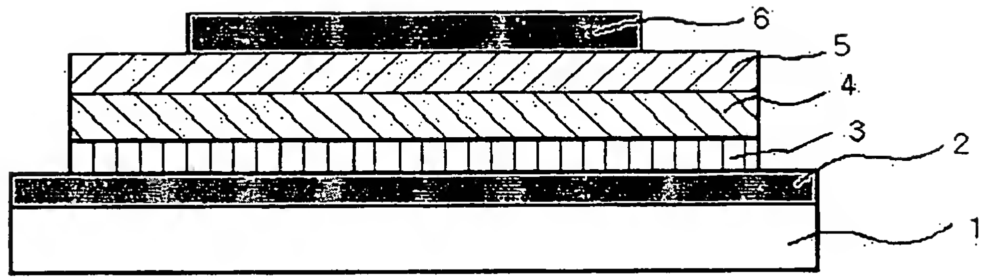
a semitransparent electrode deposited on the transparent substrate;

an emissive layer made of organic EL material, positioned on the semitransparent electrode;

an hole-injecting layer made of single-anion conductors, positioned on the emissive layer; and,

a metal electrode deposited on the hole-injecting layer.

Fig. 1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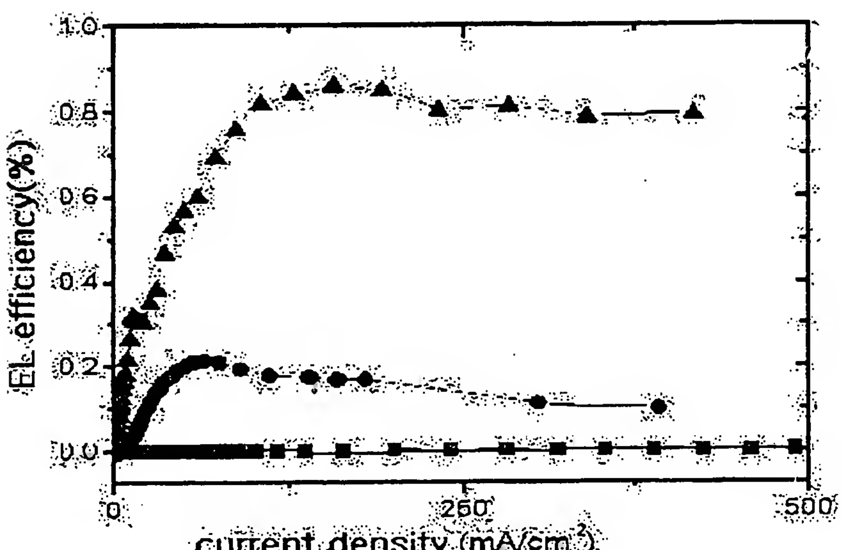
5
4
3
2
1

Fig. 2


1.0
0.8
0.6
0.4
0.2
0.0
EL efficiency(%)
0
250
500
current density (mA/cm^2)

INTERNATIONAL SEARCH REPORT

International application No.
PCT/KR01/00535

A. CLASSIFICATION OF SUBJECT MATTER

IPC7 H05B 33/14 , H05B 33/20

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimun documentation searched (classification system followed by classification symbols)

IPC7 H05B 33/14, H05B 33/20

Documentation searched other than minimun documentation to the extent that such documents are included in the fileds searched

Korean patents and applications for invention since 1975
Korean utility models and applications for utility models since 1975

Electronic data base consulted during the intertnational search (name of data base and, where practicable, search trerms used)

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
"A"	JP 10-308277 A (NIPPON ELECTRIC CO) 17.NOV.1998 (WHOLE DOCUMENT)	1-9, 12-17
"A"	JP 11-233262 A (AIMES CO) 27.AUG.1999. (WHOLE DOCUMENT)	1-9, 12-17
"A"	US 6,030,715 A (UNIVERSITY OF SOUTHERN CA.) 29.FEB. 2000 (WHOLE DOCUMENT)	1-9, 12-17

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Date of the actual completion of the international search	Date of mailing of the international search report
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INTERNATIONAL SEARCH REPORT

Information on patent family members

International application No.

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